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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF	:	
Hisao IKEDA et al	:	GROUP ART UNIT: 1624
SERIAL NO.: 09/973,766	:	EXAMINER: V. BALASUBRAMANIAN
FILED: October 11, 2001	:	

FOR: METHOD FOR REDUCING AN ORGANIC SOLVENT REMAINING IN BETA-FORM TRIS-(2,3-EPOXYPROPYL)-ISOCYANURATE CRYSTALS

DECLARATION UNDER 37 CFR 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes Hisao Ikeda who deposes and says that:

1. I am one of the co-inventors of the above-identified application.
2. I have worked on the following experiments.

Experiment 1 (corresponding to Example 6 of the specification)

Step (A): Into a flask equipped with a stirrer, a thermometer, a continuously dropping apparatus and an apparatus to concentrate an azeotropic vapor of epichlorohydrin and water under reduced pressure and to return only epichlorohydrin to the reaction system, 774 g (6 mols) of cyanuric acid, 8,328 g (90 mols) of epichlorohydrin and 213 g of a tetramethylammonium chloride aqueous solution having a concentration of 15.5 wt%, were added and refluxed with stirring at from 89 to 120°C for 5 hours to carry out the reaction. Then, the temperature of the reaction system was cooled to 50°C, and 1,536 g of a 50 wt% sodium hydroxide aqueous solution was added and reacted for 6 hours under a reduced pressure of from 100 to 60 Torr with stirring while maintaining the temperature at 50°C for dehydrochlorination. Then, formed sodium chloride was dissolved by an addition of 3,600 g of water for washing, followed by liquid separation, and 1,200 g of a 5 wt% sodium dihydrogen phosphate aqueous solution was further added for washing, whereby sodium hydroxide used in an excess amount was neutralized, followed by washing with 4,800 g of water.

Step (B): To 1,600 g of tris-(2,3-epoxypropyl)-isocyanurate obtained by evaporating the reaction solution to dryness at 120°C under 2 Torr, 1,600 g of acetonitrile was added.

Step (C): The temperature was raised to 75°C to completely dissolve the solid content, then the liquid was cooled to 14°C over 7 hours to precipitate β-form tris-(2,3-epoxypropyl)-isocyanurate crystals, which were collected by filtration.

Step (D): The obtained crystals were washed with 600 g of methanol, followed by filtration.

The obtained cake was dried at 80°C under a reduced pressure of 5 Torr for 4 hours to obtain β-form tris-(2,3-epoxypropyl)-isocyanurate crystals in a yield of 288 g. The obtained crystals had amounts of remaining acetonitrile and

epichlorohydrin of 1,000 ppm and 60 ppm, respectively, an epoxy equivalent of 102 g/eq., an amount of hydrolyzable chlorine of 240 ppm, a total chlorine amount of 260 ppm, a content of α-form tris-(2,3-epoxypropyl)-isocyanurate extractable by methanol of 0.5 wt% and a content of α-form tris-(2,3-epoxypropyl)-isocyanurate not extractable by methanol contained in the interior of the crystals of 9.0 wt%. The crystals had a melting point of from 149 to 156°C, an average particle size of 60 μm, and they were white crystals. Further, the crystals were dried in a nitrogen stream at 130°C under a reduced pressure of 10 Torr by a rotary evaporator for 24 hours to obtain β-form tris-(2,3-epoxypropyl)-isocyanurate crystals wherein the amounts of remaining acetonitrile and epichlorohydrin were 220 ppm and 40 ppm, respectively and a remaining total chlorine amount of 45 ppm.

Experiment 2 (Comparative Example)

Step (A): Into a flask equipped with a stirrer, a thermometer, a continuously dropping apparatus and an apparatus to concentrate an azeotropic vapor of epichlorohydrin and water under reduced pressure and to return only epichlorohydrin to the reaction system, 774 g (6 mols) of cyanuric acid, 8,328 g (90 mols) of epichlorohydrin and 213 g of a tetramethylammonium chloride aqueous solution having a concentration of 15.5 wt%, were added and refluxed with stirring at from 89 to 120°C for 5 hours to carry out the reaction. Then, the temperature of the reaction system was cooled to 50°C, and 1,536 g of a 50 wt% sodium hydroxide aqueous solution was added and reacted for 6 hours under a reduced pressure of from 100 to 60 Torr with stirring while maintaining the temperature at 50°C for dehydrochlorination. Then, formed sodium chloride was dissolved by an addition of 3,600 g of water for washing, followed by liquid separation, and 1,200 g of a 5 wt% sodium dihydrogen phosphate aqueous solution was further added for washing, whereby sodium hydroxide used in an excess

amount was neutralized, followed by washing with 4,800 g of water.

Step (B): To 1,600 g of tris-(2,3-epoxypropyl)-isocyanurate obtained by evaporating the reaction solution to dryness at 120°C under 2 Torr, 2,400 g of dichloroethane was added.

Step (C): The temperature was raised to 73°C to completely dissolve the solid content, then the liquid was cooled to 25°C over 6 hours to precipitate β-form tris-(2,3-epoxypropyl)-isocyanurate crystals, which were collected by filtration.

Step (D): The obtained crystals were washed with 600 g of methanol, followed by filtration.

The obtained cake was dried at 80°C under a reduced pressure of 5 Torr for 4 hours to obtain β-form tris-(2,3-epoxypropyl)-isocyanurate crystals in a yield of 264 g. The obtained crystals had amounts of remaining dichloroethane and epichlorohydrin of 1,400 ppm and 50 ppm, respectively, an epoxy equivalent of 102 g/eq., an amount of hydrolyzable chlorine of 220 ppm, a total chlorine amount of 1,240 ppm, a content of α-form tris-(2,3-epoxypropyl)-isocyanurate extractable by methanol of 0.5 wt% and a content of α-form tris-(2,3-epoxypropyl)-isocyanurate not extractable by methanol contained in the interior of the crystals of 8.0 wt%. The crystals had a melting point of from 149 to 156°C, an average particle size of 60 μm, and they were white crystals. Further, the crystals were dried in a nitrogen stream at 130°C under a reduced pressure of 10 Torr by a rotary evaporator for 24 hours to obtain β-form tris-(2,3-epoxypropyl)-isocyanurate crystals wherein the amounts of remaining dichloroethane and epichlorohydrin were 180 ppm and 20 ppm, respectively, and a remaining total chlorine amount of 150 ppm.

Note:

In Experiment 1:

Epichlorohydrin amount (ppm) = epichlorohydrin amount in the formed crystal

Hydrolyzable chlorine amount (ppm) = chlorine amount of chlorohydrin compound decomposed by hydrolysis for measurement and measured by titration method

Total chlorine amount (ppm) = chlorine amount of chlorohydrin compound decomposed by combustion method for measurement and measured by titration method

In Experiment 2:

Epichlorohydrin amount (ppm) = epichlorohydrin amount in the formed crystal

Hydrolyzable chlorine amount (ppm) = chlorine amount of chlorohydrin compound decomposed by hydrolysis for measurement and measured by titration method

Total chlorine amount (ppm) = chlorine amount of dichloroethane + chlorine amount of chlorohydrin compound decomposed by combustion method for measurement and measured by titration method

* The above chlorohydrin compound includes chlorohydrin as an intermediate compound of tris-(2,3-epoxypropyl)-isocyanurate and a hydrolysate of epichlorohydrin.

(Consideration)

As evident from the experimental data, Experiment 1 (corresponding to Example 6 of the present specification) proves that when such a chlorine-free solvent as defined in present Claim 1 is used, a hydrolyzable chlorine amount (a

remaining amount of epichlorohydrin used in step A) and a total chlorine amount (calculated on the basis of epichlorohydrin used in step A) are small.

On the other hand, Experiment 2 (Comparative Example) proves that when a chlorine-containing solvent such as dichloroethane is used, a total chlorine amount (a remaining amount of dichloroethane used in step B) is much larger than that of Experiment 1 although the hydrolyzable chlorine amount (a remaining amount of epichlorohydrin used in step A) is not significantly different between the two experiments.

3. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

4. Further declarant saith not.

Respectfully submitted,

September 7, 2004
Date

Hisao Ikeda
Hisao Ikeda